

## Evaluation of Uncertainty in Constituent Input Parameters for Modeling the Fate of RDX

by Mark S. Dortch

**ABSTRACT:** The Training Range Environmental Evaluation and Characterization System (TREECS<sup>TM</sup>) was reapplied for a previously validated model of Demolition Area 2 of the Massachusetts Military Reservation (MMR) on Cape Cod. The purpose of the exercise was to evaluate the importance of chemical-specific model input parameters, the impacts of their uncertainty, and the potential benefits of having improved estimates from the Environmental Fate Simulator (EFS). RDX migration to groundwater was the focus of the modeling. There were two chemical-specific inputs for RDX that were determined to be sensitive with relatively high uncertainty: these included the soil-water linear partitioning distribution coefficient ( $K_d$ , L/kg) and the half-life, which is related to the degradation rate. Testing revealed that results were only sensitive to  $K_d$  and half-life for the vadose zone. Results indicated that RDX degradation is very slow for the aerobic media of this site, resulting in half-life on the order of 100 years. The EFS does not presently have the capability to estimate degradation rates. Results also indicated that the organic carbon-to-water partitioning coefficient  $K_{oc}$  can be used to estimate  $K_d$ , and log  $K_{oc}$  of RDX is on the order of 1.0. The EFS can provide estimates of  $K_{oc}$  for organic chemicals. The EFS values provided for log  $K_{oc}$  of RDX were 1.72 and 1.95.

**OBJECTIVE:** TREECS<sup>TM</sup> (<u>http://el.erdc.usace.army.mil/treecs/</u>) was developed by the U.S. Army Engineer Research and Development Center (ERDC) for the Army. TREECS<sup>TM</sup> has varying levels of capability to forecast the fate of munitions constituents (MC), such as high explosives (HE), metals, and other contaminants found within and transported from firing/training ranges (and other types of source zones) to surface water and groundwater. The overall purpose of TREECS<sup>TM</sup> is to provide environmental specialists with tools to assess the potential for MC and contaminant migration into surface water and groundwater systems and to assess range management strategies for ensuring protection of human health and the environment. TREECS<sup>TM</sup> is being applied as part of the Environmental Security Technology Certification Program (ESTCP) work unit ER-201435 entitled "Field Demonstration and Validation of the Training Range Environmental Evaluation and Characterization System (TREECS<sup>TM</sup>) and Environmental Fate Simulator (EFS) for the Risk Assessment of Contaminants on DoD Ranges." The EFS provides improved estimates of chemical-specific fate parameters required for fate and transport models such as those within TREECSTM. The objective of the present investigation is to evaluate the importance of chemical-specific model input parameters, the impacts of their uncertainty, and the potential benefits of having improved estimates from EFS.

**TECHNICAL APPROACH:** Chemical-specific properties that affect constituent fate include factors such as solubility, Henry's law constant, soil and sediment partitioning distribution

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14. ABSTRACT

The Training Range Environmental Evaluation and Characterization System (TREECS???) was reapplied for a previously validated model of Demolition Area 2 of the Massachusetts Military Reservation (MMR) on Cape Cod. The purpose of the exercise was to evaluate the importance of chemical-specific model input parameters, the impacts of their uncertainty, and the potential benefits of having improved estimates from the Environmental Fate Simulator (EFS). RDX migration to groundwater was the focus of the modeling. There were two chemical-specific inputs for RDX that were determined to be sensitive with relatively high uncertainty: these included the soil-water linear partitioning distribution coefficient (Kd, L/kg) and the half-life, which is related to the degradation rate. Testing revealed that results were only sensitive to Kd and half-life for the vadose zone. Results indicated that RDX degradation is very slow for the aerobic media of this site, resulting in half-life on the order of 100 years. The EFS does not presently have the capability to estimate degradation rates. Results also indicated that the organic carbon-to-water partitioning coefficient Koc can be used to estimate Kd, and log Koc of RDX is on the order of 1.0. The EFS can provide estimates of Koc for organic chemicals. The EFS values provided for log Koc of RDX were 1.72 and 1.95.

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coefficients, and various forms of degradation in the natural environment, such as oxidation, photolysis, hydrolysis, and biodegradation. Some properties are known or can be fairly accurately estimated, such as solubility, while others — such as degradation rates — are often far more uncertain. Prior to using improved methods for estimating chemical-specific properties that affect fate, it is useful to first evaluate the uncertainty in values for chemical-specific properties. To meet this purpose, a previous application of TREECS<sup>TM</sup> was used to evaluate parameter sensitivity and the effects of highly uncertain inputs for properties. Site-specific, physically based inputs, such as meteorology, length scales, soil properties, hydrology, etc., were not considered in this analysis. The previous application of TREECS<sup>TM</sup> that was selected for study was the one for Demolition (Demo) Area 2 at Massachusetts Military Reservation (MMR) on Cape Cod, used in 2012 as part of the TREECS<sup>TM</sup> initial validation. The HE RDX was the MC of concern at this site. This validation application was revisited in the present work to evaluate the importance of chemical-specific model input parameters, the impacts of their uncertainty, and the potential benefits of having improved estimates from EFS. This Technical Note (TN) describes the results of this investigative application.

**BACKGROUND:** Demo Area 2 of MMR (Figure 1) was used for light demolition training for roughly 10 years, beginning in about 1978 and continuing until about 1988. The area was used for demolition training, not for demolition of loaded munitions; thus, non-munitions objects were blown up rather than munitions containing explosives. Range records show that the explosives used in this area were limited to blocks of C4 and TNT demolition charges. Consequently, C4, which contains RDX and plasticizers, was a prevalent explosive at this site. Some charges may not have experienced full, high-order detonation, resulting in unexploded HE residue. RDX residue from these explosives infiltrated the groundwater beneath the demolition range. Soil and groundwater concentrations of RDX were measured at the site about 10 to 15 years after demotion training had ceased. The RDX groundwater plume delineation relative to monitoring wells near Demo Area 2 is shown in Figure 2.

**VALIDATED MODEL RESULTS:** The application of TREECS<sup>TM</sup> to Demo Area 2 of MMR is described by Dortch (2012). The application focused on the fate and transport of RDX in soil, vadose zone, and groundwater with comparisons to measured RDX concentrations in soil and groundwater. The application involved applying the TREECS<sup>TM</sup> Tier 2 soil model, the Multimedia Environmental Pollutant Assessment System (MEPAS) vadose zone model, and the MEPAS aquifer model.

When the input files for the previous validation application were inspected, it was discovered that the input for vertical dispersivity in the vadose zone was off by a factor of 10. The intention was to set the value for this parameter to 1/100 of the depth of the water table below ground surface, which is 39.6 m. The dispersivity was set to 3.96 m rather than 0.396 m as intended. This correction was made, and the models were rerun. The results were similar to those obtained previously, except for a less-rounded shape for the plot of RDX concentration versus time in groundwater. Additionally, a minor change was made for the soil-water linear partitioning coefficient  $K_d$  for RDX in soil. The appropriate estimated value for  $K_d$  is 0.203 L/kg when using a measured value of 13.2 L/kg for the organic carbon-water linear partitioning coefficient  $K_{oc}$  along with silty loam with 1.7 % organic matter. These values of  $K_d$  and  $K_{oc}$  are about double the values used in the validation application in which those values were based on estimates of  $K_{oc}$  from the octanol-water partitioning coefficient,  $K_{ow}$ .

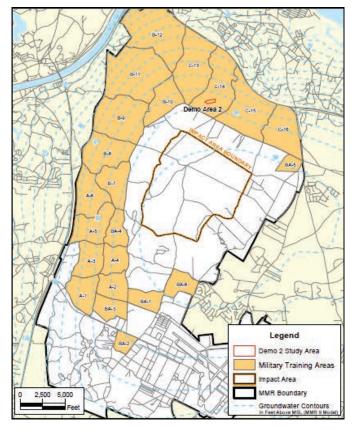


Figure 1. Demo Area 2 of MMR, Cape Cod, MA.

The model results after the correction/change are shown in Figures 3 and 4 for soil and groundwater, respectively, with comparison to the mean and range of measured concentrations. Although fairly good model agreement with observations was obtained at all wells, groundwater monitoring well MW161 was selected for comparisons. Groundwater measurements extended over a couple of years, but they all were assumed to have been collected in one year, 2003, in order to develop the mean and range of observed concentrations.

**MODEL UNCERTAINTY ANALYSIS:** As with all modeling, there are uncertainties in the prescribed model inputs. The uncertainty is greater for some inputs than others. One of the most uncertain inputs in this application is the loading rate (grams/year) of unexploded RDX residue. A value of 1.5 kg/yr was used and seemed reasonable based on the demolition charges used. However, there is no question that this loading rate is purely a rough estimate. Media concentrations are linearly proportional to the loading rate (Dortch et al. 2010).

The average size of unexploded, solid-phase RDX particles is another uncertain input that is required to compute the solid particle dissolution rate in water during precipitation. Explosives residue at this site is a result of detonations that did not fully expend all of the HE material, which is referred to as low-order detonation. The particles of low-order detonations are on the order of a centimeter in size (Pennington et al. 2005 and Taylor et al. 2004). A particle diameter of 6,000 micrometers or 0.6 cm was used for this application. Doubling this size to 1.2 cm resulted in a 48% reduction in the peak RDX groundwater concentration at MW161 from 1.31  $\mu$ g/L (or parts per billion, ppb) to 0.68 ppb.

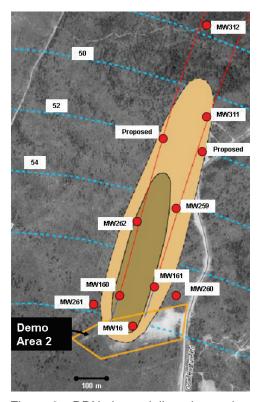


Figure 2. RDX plume delineation and monitoring wells for Demo Area 2 with groundwater contours in feet National Geodetic Vertical Datum (NGVD) (modified from AMEC Earth and Environmental (2004)).

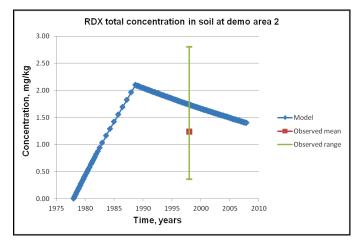


Figure 3. Computed and observed soil concentrations of RDX at Demo Area 2.

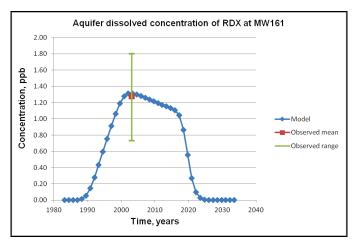


Figure 4. Computed and observed groundwater concentrations of RDX at MW161 down gradient of Demo Area 2.

The MEPAS aquifer model that is used in TREECS<sup>TM</sup> is a semi-analytical-numerical constituent transport model that includes one-dimensional Darcy advection with three-dimensional dispersion, sorption, and first-order degradation. The model solves for groundwater concentrations over time for specified well locations relative to the perceived plume centerline. Of course, the plume centerline may not be well known in most cases. Usually, the centerline is assumed to align in the primary direction of Darcy flow, or along the direction of the water table gradient, with the origin of the direction vector starting at the center of the contaminated source zone. However, the true plume centerline of flow can be different from this assumed direction; thus, there can be error in estimating the distances from the perceived plume centerline to known well locations. A variation of a few meters in the lateral distance from centerline to well location can have a minor effect in some applications, but in this application, such a variation had a substantial effect. For example, moving the lateral distance from centerline for MW161 two meters further out from 59 to 61 m resulted in a 34% decrease in peak RDX groundwater concentration at MW161 from 1.31 ppb to 0.87 ppb.

An additional set of physical parameters with a high degree of uncertainty and which substantially affect results are the dispersivity factors for groundwater transport. There is a factor for each of three dimensions, longitudinal, lateral or transverse, and vertical, where values of 1.09, 0.109, and 0.00198 m, respectively, were used in this application. These values, which were based on field tracer studies, were used in previous modeling of this site (Dortch et al. 2007 and Dortch 2012). In most applications, such data are not available, and default estimates must be made for dispersivity. Default dispersivities are based on prespecified factors and longitudinal distance from the source to the well. For MW161, the longitudinal, transverse, and vertical default dispersivities are 19.8, 6.53, and 0.0495 m, respectively, which are more than an order of magnitude larger than those used. Using the default dispersivities resulted in a 66% decrease in peak RDX groundwater concentration at MW161 from 1.31 ppb to 0.44 ppb. This sensitivity and uncertainty indicates the need for improved methods for estimating dispersivity when field tracer data are nonexistent.

Model results were sensitive to all model inputs discussed above, which were substantially uncertain, site specific, and physically based. There are other site-specific inputs for the three TREECS<sup>TM</sup> models used in this application, such as soil texture/composition, meteorology, etc., but most of those inputs are much better known or estimated; such inputs are not discussed here. None of the uncertain inputs discussed above are chemical specific. The remainder of this section focuses on the two chemical specific inputs that were determined to be sensitive with relatively high uncertainty; these included the soil-water linear partitioning distribution coefficient (K<sub>d</sub>, L/kg) and the half-life (which is related to the degradation rate) in the vadose zone. Solubility, another chemical-specific input, affects the particle dissolution rate, but solubility of RDX is well known. The Henry's Law constant (HLC) is a chemical-specific input that affects volatilization, but values for it are either known or can be reliably estimated with models, such as those included in the EPI Suite software developed by the U.S. Environmental Protection Agency's Office of Pollution Prevention Toxics and Syracuse Research Corporation (<a href="http://www.epa.gov/oppt/exposure/pubs/episuitedl.htm">http://www.epa.gov/oppt/exposure/pubs/episuitedl.htm</a>). EPI Suite is also part of EFS. Furthermore, HLC for RDX is so small (6.23E-8 atm-m<sup>3</sup>/mole), there is practically no volatilization.

The half-lives of RDX in soil and groundwater are also highly uncertain inputs, but their values are expected to be high in this application. RDX does not readily degrade in aerobic systems (Speitel et al. 2001 and Hawari 2000). The soil, vadose zone, and even the groundwater at Demo Area 2 of MMR are aerobic. Half-lives of RDX on the order of years and much longer are reported for aerobic systems (Speitel et al. 2001 and Ronen et al. 2008). An RDX half-life of 100 years was used in the validated model for soil, vadose zone, and groundwater. An RDX half-life in soil of a year and higher had little to no effect on groundwater concentrations for this study site. Similarly, a half-life of a year or more in groundwater had no effect on groundwater concentrations at this site due to the relatively short travel time of about two months from the groundwater below the source zone to MW161. However, the travel time through the vadose zone from surface soil to the water table is approximately 12 years. Thus, half-lives for the vadose zone of less than 100 years have a profound effect on groundwater concentrations. Halflives greater than 100 years produced results very similar to those obtained using a 100-year half-life. For example, assuming no degradation in the vadose zone resulted in a peak groundwater concentration of RDX at MW161 of 1.43 ppb compared with 1.31 ppb for a 100year half-life, which is the value used for the validated model.

The soil  $K_d$  value for organic chemicals can be estimated within the model user interfaces based upon the organic carbon–water sorption partition coefficient  $K_{oc}$ , the soil class (e.g., silty loam), which sets the percent sand, silt, and clay, and the percent organic matter content. The surface soil class at the study site is silty loam, whereas the below-ground surface (bgs) soil class required for vadose zone and aquifer modeling is sand. The values used for organic matter content of surface soil and bgs soil were 1.7 % and 0.17 %, respectively. The recommended value of  $K_{oc}$  is 13.2 L/kg based upon a measured value in one of the TREECS<sup>TM</sup> constituent databases. These inputs result in an estimated  $K_d$  of 0.203 L/kg for surface soil and 0.024 L/kg for bgs soil; these two values were used for the revised, validated soil and vadose/aquifer models, respectively. Thus,  $K_{oc}$  was actually the uncertain input. Values of  $K_{oc}$  for RDX provided by EPI Suite were 51.7 and 89.1 L/kg. Values for  $K_{oc}$  can be estimated from  $K_{ow}$ . EPI Suite provided an estimate of  $K_{ow}$  of 4.74 mL/mL and one experimental value of 7.41 L/kg. Higher values of  $K_{oc}$  and  $K_d$  cause greater retardation of RDX transport, which results in attenuation of the groundwater concentration versus time curve, exhibiting a rounder curve that peaks later at lower concentration. Estimates for  $K_{oc}$  were

considered to vary between approximately 4.6 to 195 L/kg based on values within the TREECS<sup>TM</sup> three constituent databases and estimates using  $K_{\rm ow}$ .

Sensitivity and uncertainty (S/U) analysis is provided within TREECS<sup>TM</sup> using Monte Carlo simulation with Latin Hypercube sampling. The user specifies the uncertain input variables and the statistical distributions describing their variability. The sampled output variables are also specified. Uncertainty analysis was conducted separately for RDX half-life in the vadose zone and  $K_d$  for surface and bgs soil. These two analyses are discussed below.

The half-life of RDX in the vadose zone was treated as uncertain, with a mean value of 100 years and upper and lower bounds of 300 and 5 years, respectively. A log uniform distribution was assumed since the bounds are so large, and there is no clearly known value for half-life; thus, values should receive equal consideration within the entire range of values. The Monte Carlo simulation was set to 100 iterations, although results converged within 60 iterations. The model results for RDX concentration versus time at MW161 are compared with the mean and range of observed concentration as shown in Figure 5. This plot is similar to that shown in Figure 4, except that the upper and lower 5 % and 95 % uncertainty confidence bands are included in the plot. The observed RDX concentrations confidence limits shown in Figure 5 tend to support the use of a rather high half-life.

The soil-RDX partition coefficients  $K_d$  for soil, vadose zone, and groundwater were treated as uncertain with a mean value of 0.203 L/kg for soil and 0.024 L/kg for vadose zone and groundwater. A log uniform distribution was used with upper and lower bounds of 0.071 and 3.0 L/kg, respectively, for soil  $K_d$ , and upper and low bounds of 0.0084 and 0.36 L/kg for  $K_d$  of the vadose zone and aquifer. These  $K_d$  values were estimated via the tools within the model user interfaces and using upper and lower bound  $K_{oc}$  values of 4.6 and 195 L/kg. The Monte Carlo simulation was run for 100 iterations. The model results for RDX concentration versus time at MW161 are compared with the mean and range of observed concentrations as shown in Figure 6, with the inclusion of the upper and lower 95% confidence limits due to uncertainty of  $K_d$  in soil and groundwater.

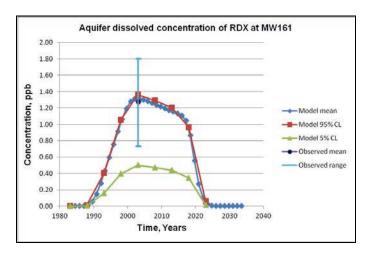


Figure 5. Computed and observed groundwater concentrations of RDX at MW161 down gradient of Demo Area 2 with upper and lower confidence bands for uncertainty on RDX half-life.

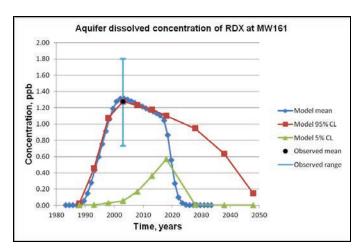


Figure 6. Computed and observed groundwater concentrations of RDX at MW161 down gradient of Demo Area 2 with upper and lower confidence bands for uncertainty on RDX K<sub>d</sub> in soil and groundwater.

The lower confidence limit in Figure 6 is much farther from the model mean (validation) than the upper limit due to the much longer transit time through the vadose zone associated with high  $K_d$  values. The longer transit time allows for greater degradation of RDX. The confidence limits in computed soil concentrations of RDX, which are not presented, were very close to the model validation result shown in Figure 3, indicating variations in the soil  $K_d$  had a minor effect on model results for this application. An additional uncertainty run was made where only the  $K_d$  value for vadose zone was treated as uncertain. The confidence limits for this run were very similar to those shown in Figure 6, thus reinforcing the conclusion that model results are sensitive to vadose zone  $K_d$  values but insensitive to surface soil and groundwater values for this application.

**CONCLUSIONS:** The application of TREECS<sup>TM</sup> demonstrated that model predictions of RDX fate can be quite sensitive to two chemical-specific inputs that are generally not well known: degradation rate (or half-life) and the soil-water partitioning distribution coefficient  $K_d$ . Values for  $K_d$  are more readily estimated than degradation rate; thus,  $K_d$  is less uncertain than half-life.

Uncertainty analysis and observed data indicated that RDX can have a long half-life (slow degradation rate), on the order of 100 years, in the vadose zone and aquifer at MMR, which are aerobic. For anaerobic systems, much faster degradation is expected. Methods for estimating degradation rates are nonexistent or poorly established, resulting in greater uncertainty in model predictions. EFS presently does not predict degradation rates.

For this application, model results were sensitive to  $K_d$  values for the vadose zone and insensitive to  $K_d$  values for soil and groundwater. This result is due to the long transit time through the vadose zone compared to soil residence time and groundwater transit time from water table entry to observation well. Higher  $K_d$  values in the vadose zone retard RDX movement, consequently allowing more time for the slow degradation rates to have an effect on RDX reduction. Uncertainty results coupled with model comparisons with observed data support a vadose zone  $K_d$  value of 0.024 L/kg, which is consistent with a  $K_{oc}$  for RDX of 13.2 L/kg, or log  $K_{oc}$  of order 1.0. Values of  $K_{oc}$  for RDX provided by EPI Suite were 51.7 and 89.1 L/kg, or log  $K_{oc}$  of 1.72

and 1.95. Although these values for  $K_{oc}$  of RDX are higher than the recommended value for this site, EFS, which includes EPI Suite, can be useful for estimating  $K_{oc}$ , and thus for estimating  $K_d$  values for organic chemicals — including explosives, propellants, and other organic contaminants. However, EFS does not have a means of estimating  $K_d$  for metals because values are highly dependent on local geochemistry. Overall, it is concluded that EFS will provide enhanced utility for applying TREECS<sup>TM</sup> by furnishing estimates for most of the chemical-specific properties required for TREECS<sup>TM</sup> application.

POINT OF CONTACT AND ACKNOWLEDGEMENT: For additional information, contact Dr. Mark Dortch, (601) 634-3517, <u>Mark.S.Dortch@usace.army.mil</u>. As mentioned previously, TREECS<sup>TM</sup> is being applied as part of the Environmental Security Technology Certification Program (ESTCP) work unit ER-201435 entitled "Field Demonstration and Validation of the Training Range Environmental Evaluation and Characterization System (TREECS<sup>TM</sup>) and Environmental Fate Simulator (EFS) for the Risk Assessment of Contaminants on DoD Ranges." This technical note should be cited as follows:

Dortch, M. S. 2015. Evaluation of uncertainty in constituent input parameters for modeling the fate of RDX. ERDC/EL TN-15-2. Vicksburg, MS: U.S. Army Engineer Research and Development Center.

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